1. **Decay mechanisms of the Mullin’s model**

**Viscous Flow:**

\[
\left( \frac{\partial W}{\partial t} \right)_{Vis} = -k_{Vis} \left( \frac{2\pi}{\lambda} \right) W = - \left( \frac{\gamma}{2\eta} \right) \left( \frac{2\pi}{\lambda} \right) W
\]

The viscous flow term is based on the Navier Stokes equation for incompressible fluids.

**Evaporation condensation, E1:**

\[
\left( \frac{\partial W}{\partial t} \right)_{E1} = -k_{E1} \left( \frac{2\pi}{\lambda} \right)^2 W = - \left( \frac{p_0 \gamma \Omega^2}{(2\pi m)^2 (k_b T)^2} \right) \left( \frac{2\pi}{\lambda} \right)^2 W
\]

The E1 term applies when a film evaporates in the presence of its own vapor.

**Evaporation condensation, E2:**

\[
\left( \frac{\partial W}{\partial t} \right)_{E2} = -k_{E2} \left( \frac{2\pi}{\lambda} \right)^3 W = - \left( \frac{\rho_0 D_g \gamma \Omega^2}{k_b T} \right) \left( \frac{2\pi}{\lambda} \right)^3 W
\]

The E2 term applies when a film evaporates in the presence of an inert gas.

**Bulk diffusion:**

\[
\left( \frac{\partial W}{\partial t} \right)_{Bu} = -k_{Bu} \left( \frac{2\pi}{\lambda} \right)^3 W = - \left( \frac{D_v \gamma \Omega^2}{k_b T} \right) \left( \frac{2\pi}{\lambda} \right)^3 W
\]

**Surface diffusion:**

\[
\left( \frac{\partial W}{\partial t} \right)_{S} = -k_{S} \left( \frac{2\pi}{\lambda} \right)^4 W = - \left( \frac{D_s \gamma \Omega^2}{k_b T} \right) \left( \frac{2\pi}{\lambda} \right)^4 W
\]
2. Details on the experimental section

2.1 Sample thickness

Figure S1. SEM image of an amorphous film cross-section. The analysis of three independent samples resulted in an average film thickness of 9.3 ± 2.0 µm (n=3, AV ± SD).
3. Results and Discussion

3.1 Determination of drug-polymer solubility using the Flory-Huggins model

Figure S2. Solubility of indomethacin (IMC) in Soluplus as a function of temperature. The drug-polymer solubility was predicted using the Flory-Huggins lattice theory (solid black curve) including the 95% prediction interval (dashed black curve). The corresponding melting point onsets of neat crystalline IMC and IMC-Soluplus physical mixtures are represented by (x).
Flory-Huggins model:

\[
\frac{\Delta H_m}{R} \left( \frac{1}{T_m} - \frac{1}{T} \right) = \ln (vD) + \left( 1 - \frac{1}{\lambda} \right)(1 - vD) + \chi (1 - vD)^2
\]

\[
vD = \frac{x}{\frac{x}{\rho_1} + \frac{1-x}{\rho_2}}
\]

With experimentally observed \(\Delta H_m\) (112.43 J/g) and \(T_m\) (160.05 °C) for IMC. The molar volume ratio of the drug and polymer (\(\lambda\)) was calculated as follows:

\[
\lambda = \frac{\frac{M_w1}{\rho_1}}{\frac{M_w2}{\rho_2}}
\]

With \(M_w1\) (118.000 g/mol) and \(\rho_1\) (1.16 g/cm\(^3\)) for Soluplus\(^1\) and \(M_w2\) (357.79 g/mol) and \(\rho_2\) (1.32 g/cm\(^3\)) for IMC\(^2\).
3.2 Fourier-transform infrared spectroscopy (FTIR)

Figure S3. FTIR spectra of neat amorphous indomethacin (IMC), Soluplus (SP) and supersaturated amorphous solid dispersions containing different amounts of SP from 600-4000 cm$^{-1}$.
3.3 Gordon-Taylor equation (G-T Eq.)

\[ T_g = \frac{w_1 T_{g1} + (K w_2 T_{g2})}{w_1 + (K w_2)} \]

\[ K = \frac{\rho_1 T_{g1}}{\rho_2 T_{g2}} \]

\( \rho_{IMC} = 1.32 \text{ g/cm}^3 \) (from Ref [2])

\( \rho_{SP} = 1.16 \text{ g/cm}^3 \) (from Ref [1])

**Table S1:** Comparison of calculated and experimentally observed glass transition temperatures.

<table>
<thead>
<tr>
<th>Soluplus concentration % (w/w)</th>
<th>Calculated ( T_g ) using G-T Eq.</th>
<th>Experimental ( T_g ) measured by mDSC*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
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<td>45.7</td>
</tr>
<tr>
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<td>45.1</td>
</tr>
<tr>
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<td>46.5</td>
<td>45.4</td>
</tr>
<tr>
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<td>47.3</td>
<td>45.5</td>
</tr>
<tr>
<td>7.5</td>
<td>48.1</td>
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<td>12.5</td>
<td>49.7</td>
<td>44.2</td>
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<td>52.2</td>
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<td>75.0</td>
<td>69.9</td>
<td>58.5</td>
</tr>
<tr>
<td>100</td>
<td>77.4</td>
<td>77.4</td>
</tr>
</tbody>
</table>

* mDSC method: starting temperature of -10 °C, heating rate of 3 °C/min with a temperature amplitude of 1 °C and a period of 40 s. For a more detailed description, see main manuscript section “instrumentation and methods.”
3.4 Calculation of $D_s$ and comparison with the only available other data

By the elimination procedure (see main manuscript and reference \[3\] of the manuscript) - backed up with experimental evidence (wavelength dependence) - we assign surface diffusion as the dominant mechanism of surface flattening:

$$ K = \frac{D_s \Omega^2 \nu}{k_b T} \left( \frac{2\pi}{\lambda} \right)^4 \quad \rightarrow \quad D_s = \frac{K k_b T}{\gamma \Omega^2 \nu} \left( \frac{2\pi}{\lambda} \right)^{-4} $$

Experimental observed $K$: $10^{-4.73}$ 1/s ($\pm 8.8\times10^{-6}$ 1/s) at $T = 303.15$ K and $\lambda = 1000$nm;

With the same values for $\nu \left( 0.05 \frac{N}{m^2} \right)$, $\Omega \left( 3\times10^{-28} m^3 \right)$ and $\gamma \left( 2\times10^{18} 1/m^2 \right)$ as mentioned in reference \[3\];

$D_s$ is calculated as $5.92\times10^{-15}$ m²/s; $-\log D_s$ as 14.23.

The value of $-\log D_s$ of roughly 14.5 from reference \[3\] (Figure 3 of the reference at 303K), indicates an approximate agreement with our observed decays.
Figure S4. XRPD diffractograms of initially amorphous IMC and the supersaturated amorphous solid dispersions with a SP concentration ranging from 1 % (w/w) to 5 % (w/w) after 306 days (and 0 days for neat IMC) of storage at 30 °C and 32 % rH. The dashed vertical lines highlight characteristic diffractions of alpha IMC. The alpha and gamma polymorphic forms of crystalline IMC are shown for reference.
References:

